A Benchmark of **predicting magnetic structures** using a Combination of the Cluster Multipole Expansion and LSDA

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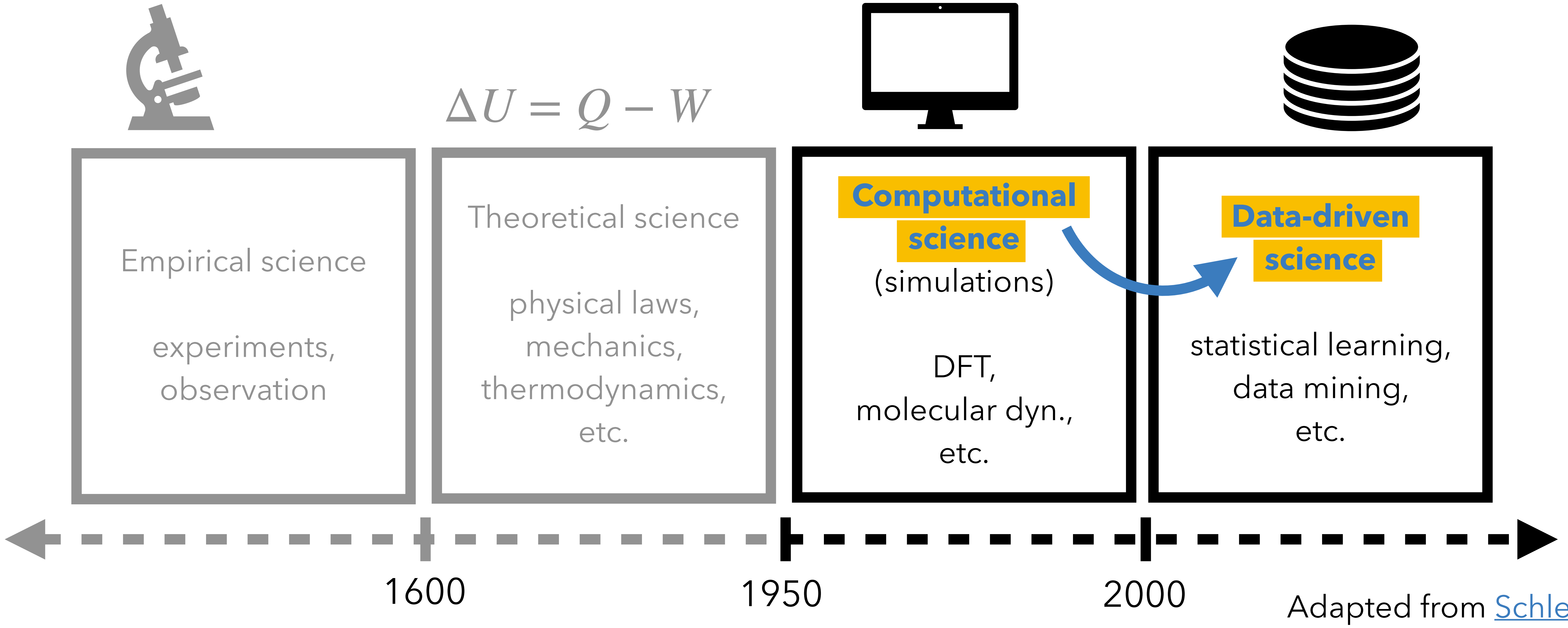
New Paradigm: Data-Driven Science



Adapted from [Schlederer et al., JPhys Mat. (2019)]

Data-driven material design:

Materials Project, AFLOW, ICSD, Clean Energy Project, …

Ex. of application:

carbon capturing [Dunstan et al., Energy Environ. Sci. (2016)] ,
battery efficiency [Qu et al., Com. Mat. Sci. (2015)],
thermoelectric materials [Toher et al., PR Mat. (2017)], …

Potential in Non-Collinear Antiferromagnetism

Symmetry-imposed shape of linear response tensors [Seemann et al., PRB (2015), Kleiner et al., PR (1966), PR (1967)]

⇒ Anomalous response in AFM is possible,

e.g. in Mn_3Sn [Nakatsuji et al., nat. (2015)], Mn_3Ge [Nayak et al., Sci. Adv. (2016)], $Mn_3Ni_{1-x}Cu_xN$ [Zhao et al., PRB (2019)], …

Advantages of non-collinear AFM:

- no stray field
- fast dynamics [Nomoto and Arita, PR Research (2019)]
- robustness against perturbations

Applications of non-collinear AFM:

- seamless and low-maintenance energy generation [Ikhlas et al., nat. phys. (2017)]
- ultrafast spintronics and robust data retention [Baltz et al., Rev. Mod. Phys. (2018)]
- fundamental understanding of magneto-transport

Computational Effort : Non-Collinear Antiferromagnetism

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Local Spin-Density Approximation (LSDA) for non-collinear magnetism [Barth and Hedin, J. Phys. C (1972)]

Challenges :

- 3 continuous degrees of freedom
- many local minima in the LSDA free energy landscape
- LSDA global minimum = true ground state ?
- magnetic unit cell = crystallographic unit cell

Goal : Predict the magnetic ground state given a compound.

Benchmark needed :

- candidate magnetic configurations ← Cluster Multipole Expansion [Suzuki et al., PRB (2019)]
- reference data ← MAGNDATA [Gallego et al., J. Appl. Cryst. (2016)]
- high-throughput LSDA ← non-coll. VASP [Hobbs and Hafner, J. Phys. Condes. Matter (2001)],
  pymatgen [Ong et al., J. Com. Mat. Sci. (2013)],
  tools on the Bilbao Crystallographic Server [Aroyo et al., Bul. Chem. Com. (2011)]

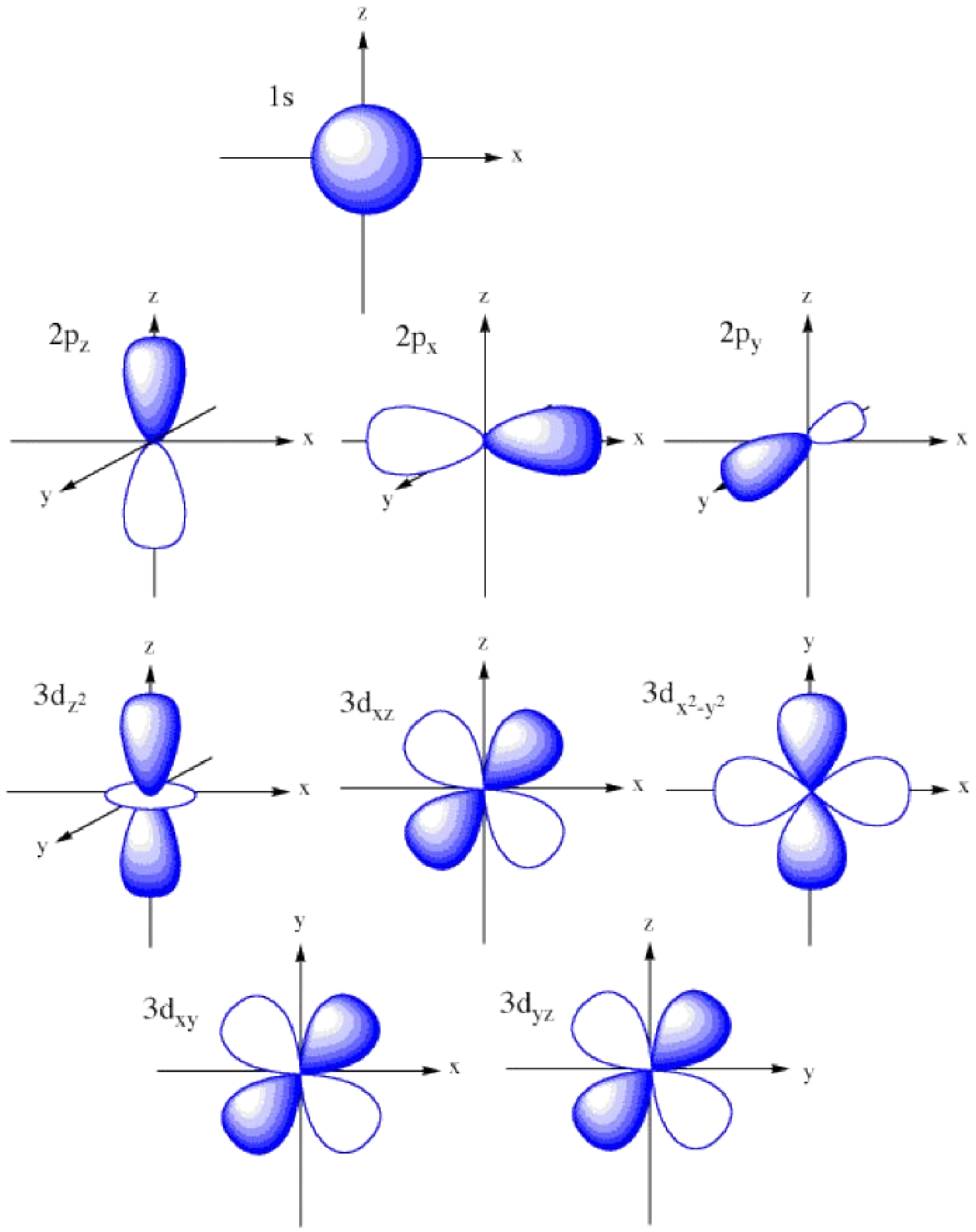
Multipole Expansion for an Electron Cloud

Scalar Poisson Equation

$$\nabla^2 \Phi(\mathbf{r}) = -4\pi\rho(\mathbf{r})$$

Scalar potential :

$$\Phi(\mathbf{r}) = \sum_{lm} \sqrt{\frac{4\pi}{2l+1}} Q_{lm} \frac{Y_{lm}}{r^{l+1}}$$



Vector Poisson Equation

$$\nabla^2 \mathbf{A}(\mathbf{r}) = -(4\pi/c)\,\mathbf{j}(\mathbf{r})$$

$$\mathbf{j}(\mathbf{r}) = c\nabla \times \mathbf{M}(\mathbf{r})$$

Vector potential :

$$\mathbf{A}(\mathbf{r}) = \sum_{lm} M_{lm} \frac{\mathbf{l} Y_{lm}}{\mathrm{i} l} \frac{1}{r^{l+1}}$$

Angular momentum :

$$\mathbf{l} = -\mathrm{i}\,(\mathbf{r} \times \nabla) \quad (l \geq 1, -l \leq m \leq l)$$

Multipole for an electron cloud :

$$M_{lm} = \sqrt{\frac{4\pi}{2l+1}} \sum_j \left(\frac{2\mathbf{l}_j}{l+1} + \sigma_j \right) \nabla \left[|\mathbf{r}_j|^l Y^*_{lm}(\varphi, \theta) \right]$$

Kusunose, JPSJ (2008)

Cluster Multipole (CMP) Expansion for Magnetic Structures

Multipole for an electron cloud:
$$M_{lm} = \sqrt{\frac{4\pi}{2l+1}} \sum_j \left(\frac{2\mathbf{l}_j}{l+1} + \sigma_j \right) \cdot \nabla \left[|\mathbf{r}_j|^l Y^*_{lm}(\varphi, \theta) \right]$$

Multipole for a point form:
$$M_{lm} = \sqrt{\frac{4\pi}{2l+1}} \sum_{i=1}^{N} \mathbf{m}_i \cdot \nabla \left[|\mathbf{r}_i|^l Y^*_{lm}(\varphi, \theta) \right]$$

Suzuki et al., PRB (2019)

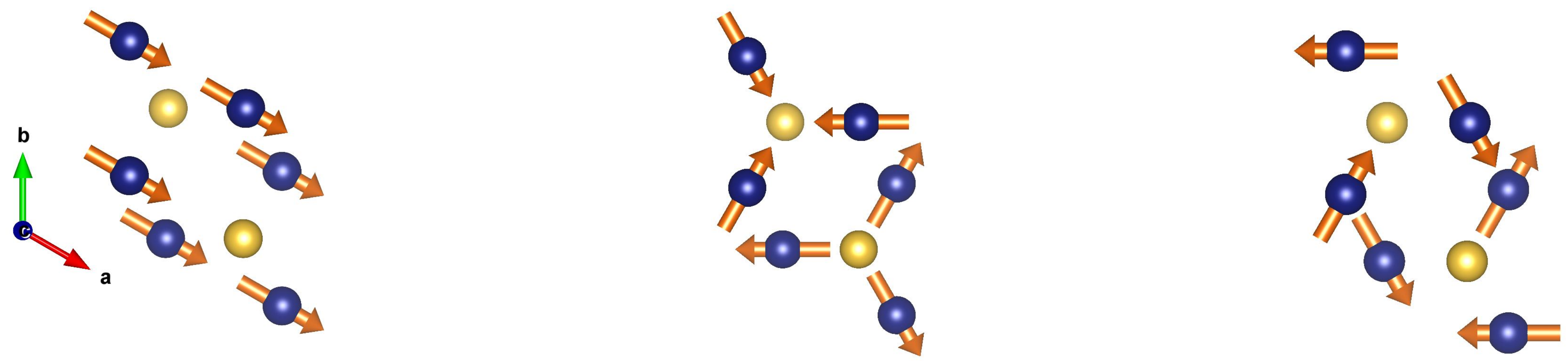
For a point group $\mathscr{P}$ a ***point form*** is a set of all symmetrically equivalent points.

N is the ***multiplicity*** of the point form.

Point form + magnetic configuration = ***(magnetic) cluster***.

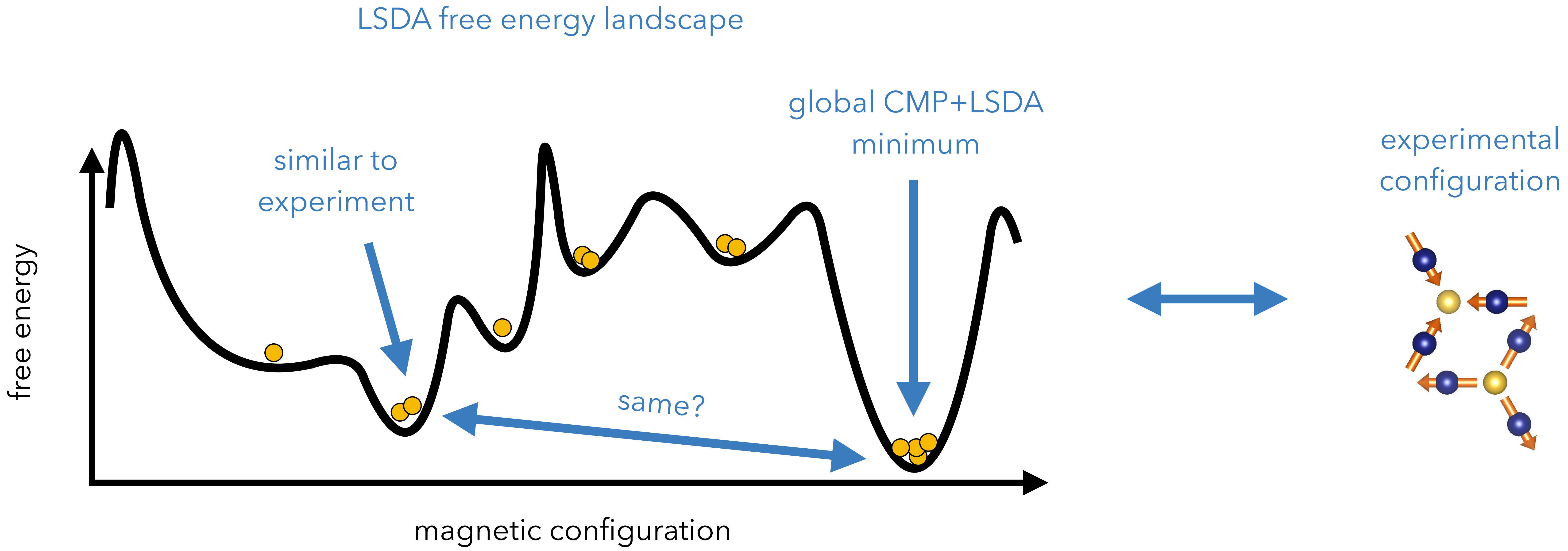
Example of CMP configurations for Mn_3Sn:

dipole | octupole | octupole



Each CMP carries a definite **order & irrep**.

High-Throughput CMP+LSDA Calculations



- Most magnetic configurations can be characterized by one or few symmetrically related CMPs.
- Experimental configuration is among the CMP+LSDA local minima.
- In about $1/3$ of the cases: CMP+LSDA global minimum = experimental configuration.

Is the Magnetic Moment per Site well-estimated?

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- No general over-/under-estimation
- No reliable prediction $\lesssim 2\mu_B$
- $\gtrsim 2\mu_B$ accurate prediction, with low precision